



Constraints on the noble gas composition of the deep mantle by bubble-by-bubble analysis of a volcanic glass sample from Iceland



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ABSTRACT

Contamination of samples by air-derived noble gases is a well-known problem in noble gas geochemistry, and, as a result, determining the true neon and argon isotopic ratios of the mantle is not straightforward. Here, we directly target individual bubbles in an Icelandic volcanic glass, DICE 11, with a 193 nm excimer laser in order to reduce air-contamination, and analyze He and Ar isotopes, plus ²²Ne abundances, on a Helix SFT mass-spectrometer. The CO₂ content of the bubbles was measured with a capacitance manometer. In addition, new He, Ne and Ar compositions obtained by crushing on similar samples (DICE 10 and DICE 11) are presented. Our analyses show that ³He/⁴He ratios are homogeneous in all the vesicles in this glass sample at 17.4 ± 0.4 Ra and are consistent with analyses by crushing. Precise ⁴⁰Ar/³⁶Ar isotopic ratios were obtained on the largest vesicles only, due to high blank contributions to the smallest vesicles, and are 8600 ± 700 , i.e. the highest values measured so far in primitive basalts from Iceland. Considering that the Ar and He isotopic compositions obtained for individual vesicles (by laser ablation) are representative of the true mantle source values, not contaminated by air, we can precisely correct the Ne isotopic analyses obtained by crushing for air contamination; the corrected ²⁰Ne/²²Ne ratios are consistent with the presence of neon-B in the sub-Icelandic mantle, which is consistent with the hypothesis that neon in the OIB source region has an irradiated meteorite origin rather than purely solar. In contrast to He and Ar isotopic compositions (which are homogeneous), the relative He–Ne–Ar–CO₂ abundances in the different bubbles are heterogeneous and show strong correlations which are compatible with equilibrium degassing of the magma. Because He and CO₂ do not fractionate during this degassing, we can precisely constrain the CO₂/³He ratio of the mantle source at $6.7 \pm 0.5 \times 10^8$, at the lower end of the MORB range. Therefore the deep mantle below Iceland is not enriched in carbon compared to the upper mantle (via recycling, for example).

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1. Introduction

In 1969, Clarke et al. and Mamyrin et al. measured ³He excesses in deep ocean waters which they related to the degassing of the Earth's mantle. Because ³He is primitive, i.e. is not produced in significant quantities in the mantle, this was the first evidence that the Earth's mantle still contains primitive noble gases. Since then, analysis of gases trapped in volcanic lavas produced by partial melting of the Earth's mantle has resulted in essential constraints on the origin of volatiles in the Earth (e.g. Craig and Lupton, 1976; Ozima and Zashu, 1983; Sarda et al., 1988), and on the evolution of the mantle volatile budget through time (e.g. Albarède, 1998; Allègre et al., 1987; Marty, 2012; Porcelli and Halliday, 2001). These studies have shown that the mantle contains solar-type neon and helium, isotopically distinct from the Earth's atmosphere (Dixon et al., 2000; Hiyagon et al., 1992; Honda et al., 1991, 1993,

1987; Kurz et al., 1985, 2009; Moreira et al., 2001; Poreda and Farley, 1992; Sarda et al., 1988; Yokochi and Marty, 2004).

However, while the noble gas composition of the upper mantle source to Mid-Oceanic Ridge Basalt (MORB) lavas is relatively well constrained (e.g. review by Graham, 2002), the volatile composition of the Ocean Island Basalt (OIB) mantle reservoir is still a subject of discussion. It has been well established that some ocean islands such as Iceland, Hawaii or Galapagos have less radiogenic helium and neon isotopic ratios (e.g. in Iceland, Breddam et al., 2000; Dixon et al., 2000; Füri et al., 2010; Harrison et al., 1999; Kurz et al., 1985; Marty et al., 1991, Moreira et al., 2001, Mukhopadhyay, 2012; Starkey et al., 2009; Trierloff et al., 2000) compared to MORBs. One of the most popular hypotheses is that the OIB mantle source is less degassed than that of MORB, and as a result the neon and helium isotopic ratios are less sensitive to U and Th decay (that produces the radiogenic elements ⁴He and ²¹Ne) through time (e.g. Allègre et al., 1983). However, this hypothesis remains controversial: for example, the noble gas content measured in OIB samples is generally lower than in MORB samples, the opposite of what would be expected for a volatile-rich mantle reservoir (the helium

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paradox – Anderson, 1998). This observation could be the result of shallow degassing processes preferentially affecting OIB lavas (e.g. Gonnermann and Mukhopadhyay, 2007; Moreira and Sarda, 2000) but it is nevertheless possible that it is instead a characteristic of the OIB mantle source (e.g. Anderson, 1998). To improve our knowledge of the mantle reservoir and to distinguish between those two possibilities, it is thus essential to measure the noble gas elemental composition in OIBs while characterizing the mechanisms of magma degassing and post-emplacment gas loss of the rocks.

In addition, noble gas analyses are generally affected by an air-like contaminant (e.g. Ballentine and Barfod, 2000). Therefore in OIBs it is rather hard to determine if lower $^{40}\text{Ar}/^{36}\text{Ar}$ ratios compared to MORBs testify for a mantle source of OIB that is less radiogenic for He, Ne but also Ar (^{40}Ar being radiogenic, while ^{36}Ar is primitive) than the upper mantle (e.g. Graham, 2002; Tieloff and Kunz, 2005), or if they are the result of a higher atmospheric contamination of OIB samples.

Although Ne is also subject to atmospheric contamination, it is possible to isotopically distinguish between mantle and air: neon contains two stable isotopes, ^{20}Ne and ^{22}Ne , that are not produced in significant quantities in the Earth's mantle (Yatsevich and Honda, 1997) i.e. they are inherited from the Earth's initial volatile budget. In a $^{21}\text{Ne}/^{22}\text{Ne}$ vs. $^{20}\text{Ne}/^{22}\text{Ne}$ diagram, the compositions of volcanic samples of mantle origin lie along mixing lines between the atmosphere ($^{20}\text{Ne}/^{22}\text{Ne}$ and $^{21}\text{Ne}/^{22}\text{Ne}$ ratios of 9.8 and 0.029, respectively) and mantle-derived components having higher $^{20}\text{Ne}/^{22}\text{Ne}$ ratios and more radiogenic $^{21}\text{Ne}/^{22}\text{Ne}$ ratios.

In addition, different noble gas carriers in the solar system, such as presolar grains, chondrites or the sun have distinct $^{20}\text{Ne}/^{22}\text{Ne}$ ratios. Therefore the value of the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio in the Earth's mantle is a direct constraint on the nature and proportion of the different components that contributed to the neon budget on Earth, and beyond to the budget of noble gases in general. MORB samples are characterized by $^{20}\text{Ne}/^{22}\text{Ne}$ ratios between the air-value and 12.5 (e.g. Moreira et al., 1998; Sarda et al., 1988). OIBs show the same feature, but in some samples values higher than 12.5 have been measured, notably a tholeiitic glass from Iceland (DICE sample) in which Tieloff et al. (2000) and Mukhopadhyay (2012) measured 12.85 ± 0.31 and 12.88 ± 0.06 , respectively (all errors reported in this manuscript are 1σ); elsewhere, Valbracht et al. (1997) measured 13.1 ± 0.35 and 12.9 ± 0.2 in olivine and in a volcanic glass, respectively from Loihi volcano (Hawaii); Yokochi and Marty (2004) measured values between 12.5 and 13 in hand-picked magnetites of a xenolith sample from Kola Peninsula, the two highest values being 12.93 ± 0.12 and 13.04 ± 0.2 . Heber et al. (2012) calculated a $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 13.36 ± 0.09 for the sun, by correcting the $^{20}\text{Ne}/^{22}\text{Ne}$ of the solar wind of 13.8–14 measured in Genesis targets (Grimberg et al., 2008; Pepin et al., 2012), from mass dependant isotopic fractionation during solar wind formation. Thus the highest $^{20}\text{Ne}/^{22}\text{Ne}$ ratios available in the literature for terrestrial samples remain lower than the solar composition ($^{20}\text{Ne}/^{22}\text{Ne} = 13.36 \pm 0.09$, Heber et al., 2012). However, because of the air contamination issue ($^{20}\text{Ne}/^{22}\text{Ne}_{\text{air}} = 9.8$), the value of the mantle $^{20}\text{Ne}/^{22}\text{Ne}$ ratio is still speculative, and these analyses provide only lower limits for the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio in the mantle, i.e. the highest values measured so far in MORBs (12.5) and in OIBs (13.04 ± 0.2). As such, the mechanism whereby these solar volatiles were incorporated into the mantle is still a subject of discussion. Several authors defend the hypothesis that neon could have been incorporated in the early Earth by capture of the protosolar nebula and dissolution in a magma ocean (e.g. Marty, 2012). This requires an early accretion of the planet Earth when the protosolar nebula was still present. $^{20}\text{Ne}/^{22}\text{Ne}$ ratios in the mantle source close to the ratio of the sun would validate this hypothesis. By contrast, lower $^{20}\text{Ne}/^{22}\text{Ne}$ ratios in the mantle, close to the value of the implanted solar wind (called neon B), would instead be compatible with the hypothesis that neon was incorporated during Earth formation by accretion of dust/chondrites containing implanted solar wind (Raquin et al., 2008; Tieloff et al., 2000), which is characterized by

$^{20}\text{Ne}/^{22}\text{Ne}$ ratios between 12.5 and 12.9, as analyzed in lunar regoliths exposed to the solar wind (Black, 1972; Eberhardt et al., 1970). Note that all the literature data mentioned above are also compatible within 1 sigma error with a neon B component, if a range of 12.5–12.9 for the implanted solar wind (coupled with sputtering) is maintained. If Ne-B (implanted solar wind) was indeed trapped in the accreting Earth, then this implies that the protosolar nebula was no longer present at the time of Earth accretion, and thus dust was not shielded from solar wind, particularly in the inner solar system. Alternatively, subduction to the mantle of atmospheric neon or of chondritic material containing presolar grains (neon A end-member with $^{20}\text{Ne}/^{22}\text{Ne} \sim 8.5$) could have lowered the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of the mantle. To distinguish between these different scenarios, decreasing the air contamination when analyzing OIB samples is a priority in order to measure the “real” mantle end-member value.

Here we combine step crushing analyses with analyses of individual vesicles by laser ablation in an Icelandic OIB sample, to 1) better constrain the degassing and post-eruptive processes affecting the noble gas contents and compositions, and 2) determine the uncontaminated $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{20}\text{Ne}/^{22}\text{Ne}$ ratios of the local mantle source. The analysis of individual vesicles by laser ablation can conclusively distinguish between equilibrium and disequilibrium degassing processes, and can identify potential post-emplacment helium loss (Burnard, 1999a; Colin et al., 2013). In addition, by selectively analyzing individual vesicles, the technique efficiently diminishes atmospheric contamination from the glass and uncontaminated $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of the source can be obtained (Raquin et al., 2008). By assuming that the Ar and He isotopic compositions obtained from individual vesicles are representative of the true mantle source values, i.e. not air-contaminated, we then use these to precisely correct the Ne isotopic analyses obtained by crushing for air contamination.

2. Sample and analytical technique

In Iceland, recent volcanism has occurred along two types of so-called neovolcanic zones: the axial rift zones and the off-rift volcanic zone. The axial rift zone crosses the island from the southwest to northwest. In the southwest Iceland, the rift zone is subdivided into the western and eastern rift zones (WRZ and ERZ). The samples studied here are pieces of a volcanic glass that was sampled in Iceland in a quarry at the southern end of the Miðfell volcanoes in the Hengill fissure swarm, 1 km east of Lake Þingvallavatn, along the WRZ. Miðfell is a 320 m high table mountain, which resulted from subglacial fissure eruption. The volcanic glass was sampled in the southwestern part of the Miðfell unit, called Dagmálafell ($64^{\circ}10'31.9''\text{N}$, $21^{\circ}02'43.0''\text{W}$), which is the oldest and composed of picritic pillow lavas erupted in the upper part of the volcanic sequence. The lavas were subglacially erupted and contain numerous gabbroic nodules and macrocrysts of olivine, plagioclase and pyroxene (e.g. Gurenko and Sobolev, 2006). The volcanic glasses in the outer part of the pillow lavas have been studied in the past for their noble gas content (Dixon et al., 2000; Harrison et al., 1999; Mukhopadhyay, 2012; Tieloff et al., 2000) because the glasses are not altered, and their compositions are not highly fractionated compared to the expected composition of the mantle. Indeed, $^4\text{He}/^{40}\text{Ar}^*$ ratios measured in these samples are between 1 and 3, the values expected in the mantle for reasonable mantle K/U ratios (Arevalo et al., 2009; Vlastelic et al., 2006) and incompatible element residence times (Galer and Onions, 1985). In addition these samples have unradiogenic $^3\text{He}/^4\text{He}$ and $^{21}\text{Ne}/^{22}\text{Ne}$ ratios, typical of the Iceland plume. They correspond to samples DICE 10 and DICE 11 in Harrison et al.'s (1999) study.

2.1. X-ray microtomography

A piece of DICE 11 volcanic glass was cut in a $5 \times 5 \times 10$ mm cube and imaged by X-ray microtomography at MATEIS laboratory, Lyon with a $4.5 \mu\text{m}$ resolution.

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